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Geospatial distribution of heavy metals in rice soils of northwestern Peru

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The presence of heavy metals in agricultural soils poses a threat to the development of sustainable agriculture and ensuring food security. The objective of this study was to evaluate the geospatial distribution of heavy metals in rice-growing soils within the Amojú River Basin, Jaen, Peru. Ninety-five soil samples were collected randomly, covering four altitudinal ranges, namely, very low (374–450 m above sea level (m a.s.l.)), low (450–571 m a.s.l.), medium (571–701 m a.s.l.) and high altitudes (701–1,034 m a.s.l.), and different agronomic management stages, including four phenological phases (seedling, tillering, filling and ripening) and two agricultural practices (resting and stubble burning). The evaluation was conducted via physicochemical analysis, the single-factor pollution index, and spatial interpolation techniques through the empirical Bayesian kriging (EBK) method. The results indicated that the contents of cadmium (Cd) and chromium (Cr) exceed the limits established in the Environmental Quality Standards (EQSs) for agricultural soils, with maximum Cd and Cr concentrations of 2 and 21 mg kg^{-1}, respectively, mainly in very low-altitude areas (374–450 m a.s.l.). The single-factor pollution index results indicated slight Cd contamination and intense Cr contamination. Likewise, high levels of arsenic (As), Cd, and Cr were detected in soils managed via stubble burning, reaching 2, 2, and 16 mg kg^{-1}, respectively. The highest Cr concentrations were located in the northeastern and western parts of the study area, whereas the highest Cd concentrations occurred in the northeastern and southwestern parts. These concentrations may be associated with potential contamination sources, with the use of phosphate fertilizers, water for cultivation, and soil erosion as key contributors. This study highlights the potential risk to rice productivity and crop safety, emphasizing the importance of implementing sustainable agricultural practices and monitoring strategies for heavy metals in soils associated with crops.

Keywords Agricultural soils, Chromium, Cadmium, Phenological phases, EQSs, EBK

Heavy metals are considered a global problem because of the high pollution levels that can be generated in the environment[1]. Their presence in agricultural soils poses a threat to the development of sustainable agriculture and food security[2,3]. Heavy metals typically accumulate in soil from natural sources, especially through weathering processes, volcanic eruptions, biological activities, and geochemical reactions[4,5], or due to anthropogenic activities such as wastewater discharge and/or excessive use of phosphorus fertilizers[6]. The degradation of metals in soil is difficult; however, heavy metals are easily absorbed by plant roots, leading to their accumulation in various plant organs[7]. High concentrations of metals such as aluminum (Al), iron (Fe), and copper (Cu) can cause toxicity in plants during their growth and can adversely affect the production, yield, and/or quality of crops[3,8].

Metals or metalloids can accumulate due to factors such as soil erosion or leaching[2], with water as a key pathway for their mobilization, especially in agricultural soils[9]. However, regardless of these factors, soils can become reservoirs of heavy metals[10]. Elevated concentrations of cadmium (Cd), arsenic (As), and lead (Pb) are not only toxic to plants but can also affect humans when they enter the food chain, with potential carcinogenic

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effects[2]. Therefore, metal and metalloid contamination is becoming a latent problem in agricultural soils worldwide[11,12].

A specific case is rice cultivation, as rice is highly vulnerable to the accumulation of Cd and As due to its production system in flooded soils[3]. These metals are translocated from the roots to the aerial parts of rice plants through physiological processes[7], with the phloem employed as the main transport pathway[13], and can accumulate at high levels in the grains, which poses a significant risk to human health[3,14]. This crop is very important in terms of ensuring food security and developing the economy, as rice is the second most produced cereal worldwide[14] and the second most important crop in Peru[15], with an approximate total production of 3,013,148.19 tons, of which the province of Jaén contributes 4,330.90 tons, involving a total of 270 producers, with an average yield of 8.05 t ha^{-1}, thus positioning it as the second most important crop in the province, surpassed only by coffee at the end of 2024[16]. Here, only the production area in the Amojú River basin is considered, and rice is cultivated under a flood irrigation system. Notably, chemical fertilizers are applied extensively because two planting seasons occur per year, along with traditional practices such as stubble burning, which have been reported as factors that can favor the accumulation and mobility of heavy metals in soil[17]. Despite the agricultural importance of this area, the available information on the soil quality and the possible presence of metal contaminants is limited.

Although several studies have focused on addressing heavy metal contamination in soils associated with rice cultivation[5], aiming to reduce uncertainties, minimize research costs, and identify sources of contamination[18], in some regions of Peru, most of these studies have focused on urban and/or mining areas[5], thereby neglecting agricultural areas such as rice fields in tropical regions. These limitations restrict the understanding of the spatial patterns of contamination in diverse productive contexts, such as the Amojú River basin. Within this context, the presence of heavy metals in soils associated with rice cultivation could be influenced by altitude[12], crop phenological stages, agricultural management practices, and the environmental conditions in each area[19].

Hence, the early determination of the presence of heavy metals in rice-growing soils is important. Therefore, the objective of this study was to identify and quantify the concentration of metals in the rice-growing basin of Amojú Valley, which is located in the province of Jaén, Cajamarca. To achieve this goal, (i) soil physicochemical parameters were evaluated; (ii) the degree of heavy metal contamination was calculated; (iii) the distribution of heavy metals was quantified within different altitudinal ranges and at various phenological stages of rice cultivation; and (iv) the geospatial distribution of heavy metals was determined through the interpolation of collected data to better understand the current state of heavy metal contamination and to implement soil phytoremediation strategies.

Methodology

Study area

This study was conducted in the Amojú River basin, which is located in the province of Jaen, Cajamarca, Peru, at elevations ranging from 374 to 1,034 m above sea level (m a.s.l.). The region is characterized by a semidry and warm climate, with high humidity throughout the year. The maximum temperatures range from 29 to 33 °C, whereas the minimum temperatures vary between 19 and 23 °C. The annual precipitation ranges from approximately 900 to 1,200 mm[20]. Sampling points were randomly distributed across the agricultural area used for rice cultivation in the Amojú River basin (Fig. 1).

General outline of the study methodology

A methodology was established to determine the geospatial distribution of heavy metals in agricultural soils dedicated to rice cultivation in the Amojú River basin in the lower part of the province of Jaen (Fig. 2).

Physicochemical characterization and quantification of heavy metals in soils associated with rice cultivation

Soil sample collection

To collect soil samples, 95 points were strategically and randomly distributed, thereby generating a 1 × 1 km^2 grid across the agricultural area with transitory crops (i.e., mainly rice) extracted from the map of existing uses of the Ecological Economic Zoning of Cajamarca[21]. Grid nodes were defined as sampling points, and these points were uploaded to a global navigation satellite system (GNSS) receiver to conduct field trips. During the field trips, all the points were georeferenced, and those located in urban areas and water and road networks were relocated to 100 m from these areas. To strengthen the analysis, the sampling points were classified into 4 altitudinal ranges (very low, low, medium and high). In addition, the number of sampling points employed ensured representativeness within the study area[22]. The sampling points were subsequently georeferenced with a Garmin GPS instrument and ArcGIS Pro 3.0.1 software. At each location, information was collected on the crop phenological stage (S1: seedling; S2: tillering; S3: grain filling; S4: maturation) and/or the agricultural practices applied (S5: fallow; S6: burning (land preparation for sowing)). Soil samples were collected from the surface layer (0–30 cm depth) with an auger to ensure the representativeness of the arable horizon. Approximately 1 kg of soil was extracted from each sampling point and stored in labeled polyethylene bags for subsequent laboratory analysis[23]. The samples were then sent to the Laboratorio de Suelos, Aguas y Foliares (LABSAF), Estación Experimental Agraria–Moquegua, Instituto Nacional de Innovación Agraria (INIA).

Laboratory analysis

The soil samples were dried at room temperature to preserve adequate aeration. The soil samples were then passed through a 2-mm sieve after stones, gravel and roots or similar objects were removed[23]. Six physicochemical parameters (hydrogen potential (pH), electrical conductivity (EC), organic matter (OM), organic carbon (OC), available phosphorus (P) and soil texture) were analyzed via the methods described in Table 1. Four of these

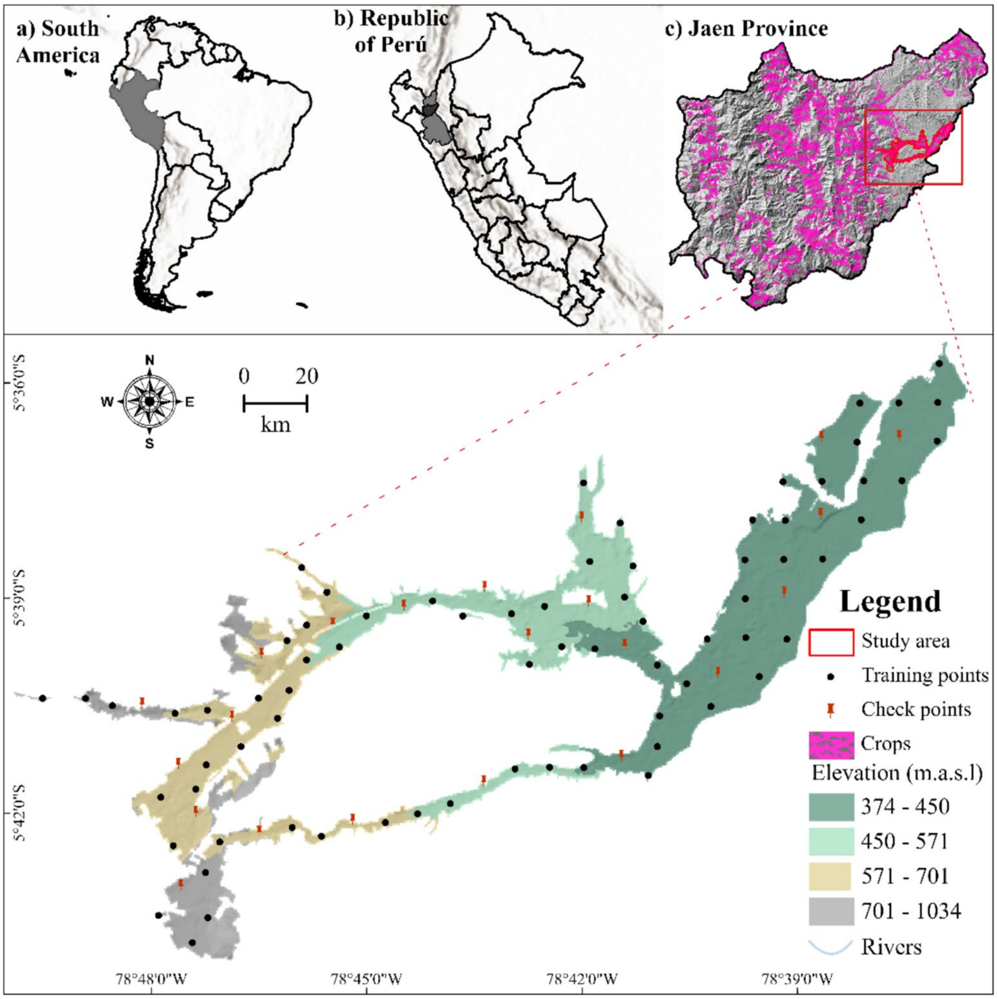


Fig. 1. Distribution of sampling points generated by the authors. Geospatially represented using Quantum GIS (QGIS) software, version 3.40.7, https://qgis.org/resources/installation-guide/#online-osgeo4w-installer. Map of current land use downloaded from the ecological and economic zoning of Cajamarca. https://geoservidor.minam.gob.pe/zee-aprobadas/cajamarca/. Elevation map downloaded from ASF Data Search Vertex https://search.asf.alaska.edu/#/.

analyses (pH, EC, OM, and textural class) were accredited by the National Accreditation Body INACAL–DA, with registration number LE—200.

For the analysis of heavy metals, the soil samples were dried in a Memmert UN260 oven at a temperature of 105 °C for 24 h[27]. Then, the soil samples were crushed in an agate mortar, sieved with a N° 10 sieve, and stored in polyethylene jars at room temperature 25. We weighed 2 g of sieved soil, added 10 mL of a nitric acid solution (HNO_3) and allowed the mixture to stand for 15 min at a temperature of 95 °C for digestion. Thereafter, we added 5 mL of concentrated HNO_3 and digested the sample for 30 min at a temperature of 95 °C. This procedure was repeated until brown vapor was no longer generated. Subsequently, 2 mL of water (H_2O) and 3 mL of hydrogen peroxide (H_2O_2) at 30% were added, and the mixture was digested for 2 h. Then, 10 mL of concentrated hydrochloric acid (HCl) was added, and the mixture was digested for 15 min at a temperature of 95 °C. Finally, the samples were filtered and gauged to 100 mL[28]. The samples were analyzed on a microwave plasma atomic spectrophotometer (S2100UV +)[29] to quantify the concentrations of cadmium (Cd), arsenic (As), lead (Pb), chromium (Cr), calcium (Ca), magnesium (Mg), iron (Fe), copper (Cu), manganese (Mn), zinc (Zn), cobalt (Co), aluminum (Al), strontium (Sr) and barium (Ba).

Quality assurance (QA) and quality control (QC)

Standard procedures were employed to ensure the reliability of the results, implementing an integral quality control system during the analysis of metals, thereby considering instrumental calibration and absorption spectra for each element analyzed (Table S1). Additionally, digestion blanks, initial calibration control standards (ICVs), continuous controls (CCVs), samples fortified with certified reference materials (CRMs), and duplicates were included. The recoveries obtained remained within acceptable ranges (95–105%), with relative deviations between duplicates smaller than 1%.

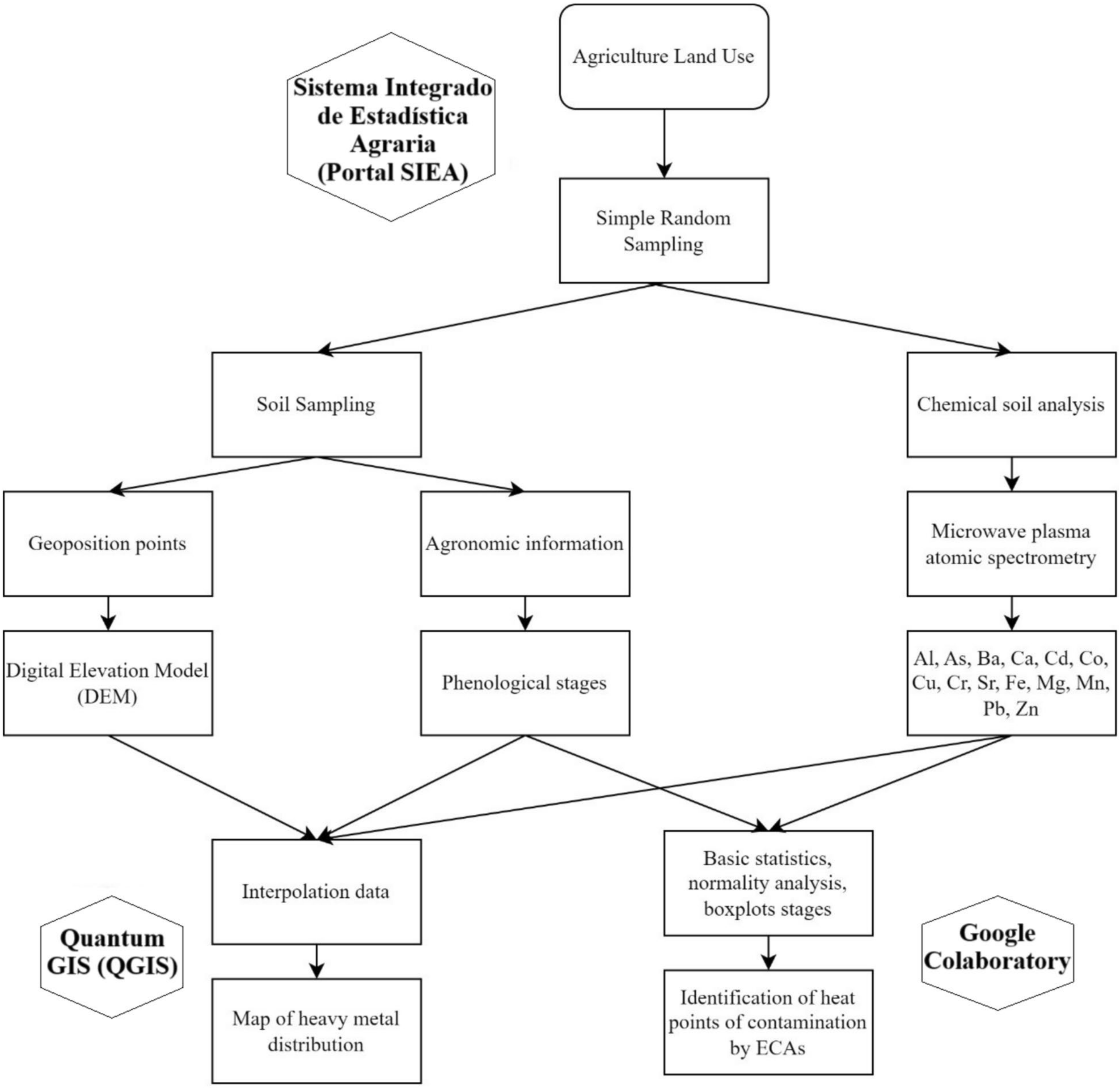


Fig. 2. Methodological scheme for determining the geospatial distribution of heavy metals.

Parameter	Unit	Method
pH	pH unit	Environmental protection agency (EPA) method 9045D[24]
EC	mS m^{-1}	ISO 11,265[25]
OM	%	AS-07 Walkley and Black. NOM-021-RECNAT-2000; second section, 2002[26].
OC	mg kg^{-1}	Walkley and Black[26]
P	mg kg^{-1}	NOM-021-RECNAT-2000; second section[26]
Soil texture	%	AS-09 Bouyoucos hydrometer method; Norma Oficial Mexicana NOM-021-RECNAT-2000[26]

Table 1. Analysis methods used for the determination of soil physicochemical parameters.

Single-factor pollution index

The single-factor pollution index (Pi) method has been commonly employed for assessing heavy metal pollution in China[30].

$$Pi = \frac{Ci}{Cbi} \tag{1}$$

where Pi denotes the contamination index of heavy metals in soil, Ci is the measured content of heavy metals, and Cbi is the detection value of the agricultural soil contamination risk for heavy metals (011–2017-MINAM) (note: 011–2017-MINAM is part of the supreme decree of the Peruvian standard Estándares de Calidad Ambiental del Suelo Agrícola, which provides limit values of select heavy metals for soil environmental quality analysis). Notably, $Pi \leq 1$ indicates the absence of contamination; $1 < Pi \leq 2$ indicates slight contamination; $2 < Pi \leq 3$ indicates moderate contamination; and $Pi > 3$ indicates intense contamination (Eq. 1).

Evaluation of the distribution of heavy metals across the different altitudinal ranges and crop stages

Spatial distribution of heavy metals in the Amojú River basin in the province of Jaen, Cajamarca

The spatial distribution of heavy metals in soils related to rice cultivation in the Amojú River basin was determined with ArcGIS Pro 3.0.1 via the EBK interpolation method and data obtained from the sampling points. These data were divided into training (80%) and validation sets (20%).

To ensure proper data interpolation with the EBK method, it is necessary to identify the data distribution beforehand, which facilitates the selection of the semivariogram model and data transformation. Therefore, to evaluate the normality of the heavy metal data, Google Collaboratory was employed. Here, the Shapiro‒Wilk and Anderson‒Darling tests were performed through the statistical module SciPy[31,32].

These tests were selected for their robustness to moderate sample sizes and their sensitivity in detecting discrepancies at distribution extremes. For the Shapiro‒Wilk test, the distribution was considered normal if the p value was smaller than 0.05 and if the statistic was close to 1. The Anderson‒Darling test results were considered normal if the statistic was less than the critical value for a normal distribution. In this case, the values were 0.549, 0.625, 0.749, 0.874, and 1.04 for significance levels of 15%, 10%, 5%, 2.5%, and 1%, respectively.

The distribution of heavy metal concentrations is shown as a frequency histogram in Fig. 3. In addition, we visually compared the curves of a theoretical normal distribution and the collected data to determine whether the data followed a normal distribution.

The distribution of the concentrations of heavy metals (Mg, Mn, Pb and Zn) resembled a normal distribution (Fig. 3). Furthermore, the normality of the data is revealed in Table 2 in greater detail on the basis of the statistical values calculated via the Shapiro‒Wilk and Anderson‒Darling tests.

The data for Mg and Mn followed a normal distribution on the basis of both the Shapiro‒Wilk and Anderson‒Darling tests. Therefore, during only the interpolation of these two parameters, the transformation parameter was set to the default value. For all nonnormally distributed elements, the EBK transformation parameter was set to corresponding empirical values, which facilitated the application of a multiplicative bias transformation with an empirical basis function in the EBK method, thus reducing the bias and increasing the interpolation quality. Finally, the K-Bessel semivariogram model was selected for its flexibility and accuracy, with 100 semivariogram simulations and a maximum of 100 points in each local model. A neighbor search in standard circular mode was performed with a minimum of 10 neighbors and a maximum of 15 neighbors, considering the computational resources.

Evaluation of the interpolation accuracy

The interpolation accuracy was measured with respect to 20% of the validation data by extracting interpolated values according to the coordinates of the validation points. In this way, a matrix of simulated and real data from the same point was established. This matrix was then used to calculate 4 spatial error analysis metrics, namely, the mean absolute error (MAE) (Eq. 2), the root mean square error (RMSE) (Eq. 3), the coefficient of determination (R2) (Eq. 4), and the concordance index (d) (Eq. 5).

$$MAE = \frac{1}{n}\sum\nolimits_{i=1}^{n} \tag{2}$$

$$RMSE = \sqrt{\frac{1}{n}\sum\nolimits_{i=1}^{n}\left(y_i - \overset{\text{‘}}{y}_i\right)^2} \tag{3}$$

$$R^2 = 1 - \frac{\sum_{i=1}^{n}\left(y_i - \overset{\text{‘}}{y}_i\right)^2}{\sum_{i=1}^{n}\left(y_i - \overline{y}\right)^2} \tag{4}$$

$$d = 1 - \frac{\sum_{i=1}^{n}\left(y_i - \overset{\text{‘}}{y}_i\right)^2}{\sum_{i=1}^{n}} \tag{5}$$

The above error metrics were calculated via Eqs 2,3,4,5 and the Numpy and Scikit-learn libraries in the Google Collaboratory environment.

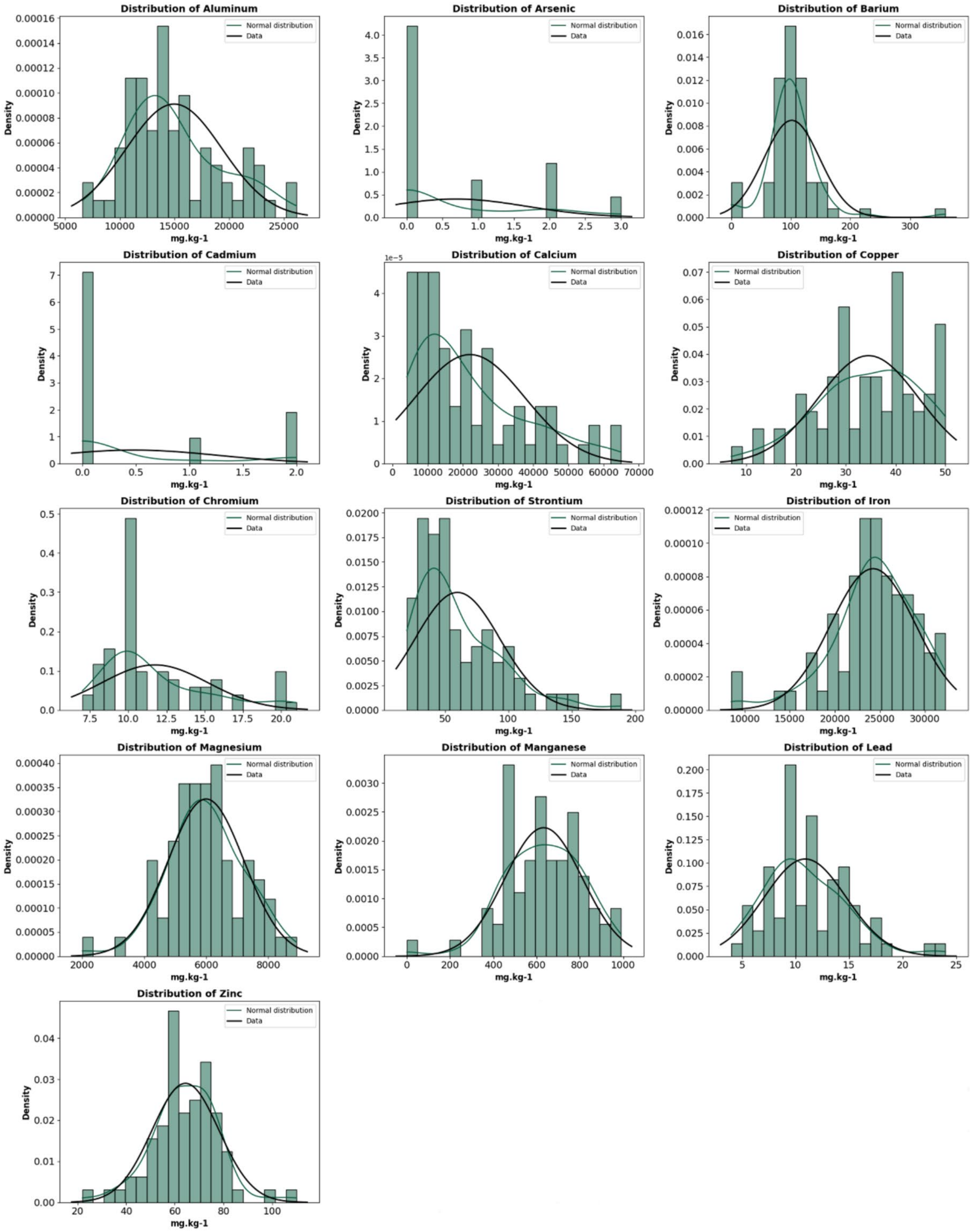


Fig. 3. Evaluation of the normality of the heavy metal concentrations in the Amojú River basin, Jaen, Cajamarca, Peru.

Analysis of the interactions between soil properties and heavy metals

The dataset was analyzed via Google Collaboratory using the Pandas library[33] to obtain basic descriptive statistics such as measures of the central tendency and dispersion. In addition, Pearson's correlation analysis was performed to determine the relationships between heavy metals and soil properties.

	Shapiro–wilk test		Anderson–darling test
Parameter	Statistic	P value	Statistic
Al	0.95	0.012	1.26
As	0.69	4.5e-11	10.25
Ba	0.78	6.1e-9	3.73
Cd	0.59	7.03e-13	14.34
Ca	0.87	4.5e-6	2.92
Co *	–	–	–
Cu	0.96	0.053	0.555
Cr	0.85	4.3e-7	4.3
Sr	0.87	2.8e-6	2.59
Fe	0.94	0.002	0.884
Mg	0.98	0.65	0.26
Mn	0.97	0.13	0.33
Pb	0.94	0.0057	0.94
Zn	0.96	0.057	0.71

Table 2. Results of the normality tests applied. * Co was analyzed, but no measurable concentrations were found.

Parameter	Unit	Median	Minimum	Maximum
pH	pH unit	7.8	5.3	8.7
EC	mS m^{-1}	21.8	7.5	146.3
OM	%	1.9	0.9	3.9
OC	mg kg^{-1}	10 986.0	5 076	22 666
P	mg kg^{-1}	11.0	4.4	65.5
Texture				
Sand	%	40.5	16.6	75.4
Clay	%	30.1	8.7	51.8
Silt	%	27.6	9.4	55.9

Table 3. Soil physicochemical parameters of the Amojú River basin. The mean, minimum and maximum values are presented for the hydrogen potential (pH), electrical conductivity (EC), organic matter (OM), organic carbon (OC), available phosphorus (P), sand, clay and silt.

Results

Physicochemical parameters of soils dedicated to rice cultivation in the Amojú River basin

In the study area, acidic (pH of 5.3) to alkaline (pH of 8.7) soils were found, and most of the samples occurred within a slightly alkaline range since the median pH is 7.8. The average EC value is 21.8 mS m^{-1}, indicating a moderate salt content in the soils associated with rice cultivation. The OM content in these soils does not exceed 4%, which indicates a low OM content. Likewise, the concentration of OC, which is related to OM, was 10,986 mg kg^{-1}, and the concentration of P was 11.0 mg kg^{-1} (Table 3).

In addition, the main soil texture is clay loam, and the proportion of sand in the soil samples was 40.5%, which could reach 75.4% at some sampling points.

Notably, with respect to the physicochemical characteristics of the soil samples, according to the four altitudinal ranges considered (very low, low, medium, and high), the pH value increased slightly with increasing elevation, with median pH values of 7.8 in the very low zone and 7.9 in the high zone. The soil pH also revealed considerable dispersion within all ranges, with outliers marked with black dots in Fig. 4a. However, different behavior was observed for the EC values. Notably, at very low elevations, the median EC value was 26.8 mS m^{-1}, and at low elevations, the median EC value was 19.0 mS m^{-1}, indicating that within these altitudinal ranges, the salt content was relatively low (Fig. 4b).

With respect to OC and OM, which are strongly determined by their nature, relatively high concentrations were obtained in the middle elevation range, with median values of 11,742.0 mg kg^{-1} for OC and 2% for OM. This finding also shows that at relatively high elevations, the OC and OM contents typically decreased (Fig. 4c and 4 d, respectively).

The concentration of P in the very low zone was high, with a median value of 11.3 mg kg^{-1}, indicating that with increasing elevation, the P concentration decreased (Fig. 4e). Finally, the soil texture within all elevation ranges was dominated by high sand contents, with a median value of 40.5%, except the high zone, in which the sand, clay and silt contents were similar, ranging from 33.8% to 35.0% (Fig. 4f).

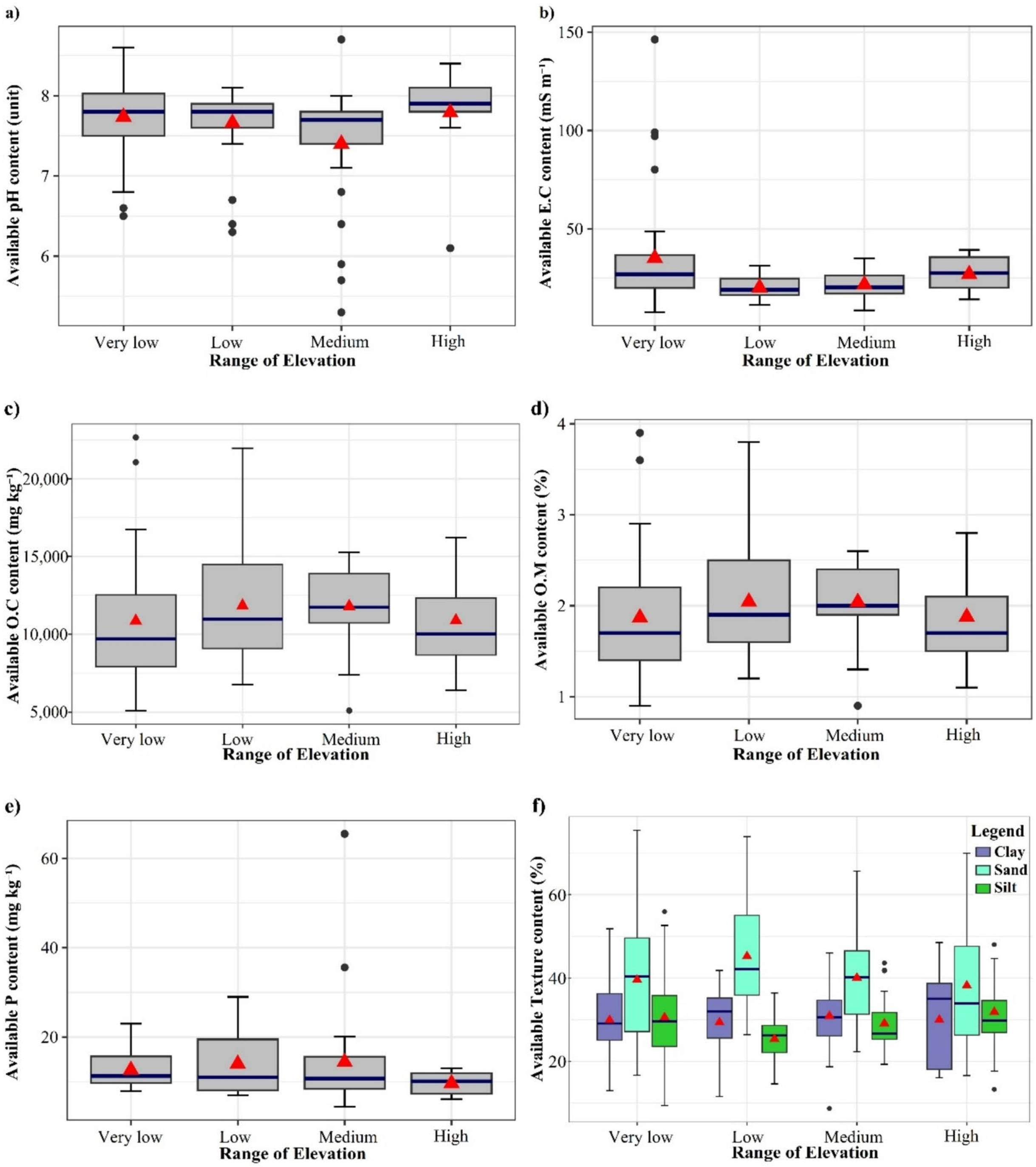


Fig. 4. Variability in physicochemical parameters according to the four altitudinal ranges. (**a**) Measured pH, (**b**) EC, (**c**) OC, (**d**) OM, (**e**) P and (**f**) texture; the altitudinal ranges are very low (374–450 m a.s.l.), low (450–571 m a.s.l.), medium (571–701 m a.s.l.) and high (701–1,034 m a.s.l.).

Concentration of heavy metals in soils associated with rice cultivation in the Amojú River basin

The concentrations of 4 heavy metals (As, Cd, Pb and Cr) and 10 essential elements in the soil samples were quantified. The maximum concentrations of Cd (2 mg kg^{-1}) and Cr (21 mg kg^{-1}) exceeded the maximum limits for agricultural soils allowed by the EPA (1.4 and 0.4 mg kg^{-1}, respectively). The concentrations of Ca, Mg, Fe, Cu, Mn, Zn, Co, Al, Sr and Ba, which are essential elements for plant growth, were also determined, whose values remained within acceptable ranges for agricultural soils (Table 4).

Variables	Unit	Median	Minimum	Maximum	EQS*
Heavy metals					
As	mg kg^{-1}	0	0	3	50
Cd	mg kg^{-1}	0	0	2	1.4
Pb	mg kg^{-1}	11	4	24	70
Cr	mg kg^{-1}	10	7	21	0.4
Elements present in soil					
Ca	mg kg^{-1}	18,645	4,049	64,924	
Mg	mg kg^{-1}	6,139	2,008	8,907	
Fe	mg kg^{-1}	24,417	7,698	32,297	
Cu	mg kg^{-1}	37	7	60	
Mn	mg kg^{-1}	659.6	239.8	1,099	
Zn	mg kg^{-1}	65	22	110	
Co	mg kg^{-1}	0	0	0	
Al	mg kg^{-1}	14,188	6,565	28,305	
Sr	mg kg^{-1}	53	20	188.9	
Ba	mg kg^{-1}	103.9	58.9	23,180	

Table 4. Concentrations of heavy metals and other elements present in soils associated with rice cultivation in the Amojú River basin in the district and province of Jaen. Note: Co was analyzed, but no measurable concentrations were found. *EQS: Environmental Quality Standard.

Altitude	Pi–Cd	Pi–Cr	Pi–Pb	Pi–As
High	0.00	25.00	0.17	0.00
Medium	0.00	25.00	0.16	0.00
Low	0.00	25.00	0.16	0.00
Very low	0.71	30.00	0.13	0.03

Table 5. Heavy metal contamination index of soils associated with rice cultivation in the Amojú River basin.

Altitude	Range	Cd	Cr	Pb	As
High	701–1,034	0	10	12	0
Medium	571–701	0	10	11	0
Low	450–571	0	10	11	0
Very low	374–450	1	12	9	1.5

Table 6. Heavy metal contents in soils associated with rice cultivation in the Amojú River basin (median in mg kg^{-1}).

Evaluation via the single-factor index method

The evaluation of heavy metal contamination in soils associated with rice cultivation revealed that 16.8% of the samples exhibited slight Cd contamination ($1 < Pi \le 2$). The Cr contamination index indicated intense contamination ($Pi > 3$). However, the heavy metals Pb and As did not exhibit contamination (Table S2).

In contrast, light Cd contamination was determined within the very low altitudinal range ($Pi = 0.71$), whereas no Pi values could be obtained within the low, medium and high altitudinal ranges. However, for Cr, the Pi values were high, with the very low, low and medium altitudinal ranges exhibiting a Pi value of 25.00, which suggests that low-elevation areas are more susceptible to contamination with this heavy metal. However, Pb and As did not exhibit significant Pi values within any of the altitudinal ranges. The highest Pi value for Pb (0.17) was obtained within the very low range, and the highest Pi value for As was obtained within the high range ($Pi = 0.03$), which do not indicate significant contamination levels according to the evaluation index for measuring the degree of accumulated contamination, as commonly employed in China (Table 5).

Distribution of heavy metals across altitudinal ranges and crop stages

Heavy metal contents in soils at different altitudes

The concentration of heavy metals in the soils of the study area varied according to altitude. In the high-altitude zone, only Cr and Pb were detected, with concentrations of 10 and 12 mg kg^{-1}, respectively. In contrast, the very low-altitude zone exhibited the presence of heavy metals such as Cd, Cr, Pb, and As, with concentrations of 1, 12, 9, and 1.5 mg kg^{-1}, respectively (Table 6).

Distribution of heavy metals in soils within the four altitudinal ranges

A distribution map of As, Cd, Pb and Cr in the four altitudinal ranges (very low, low, medium and high) was generated, and Cr (yellow bars) and Pb (red bars) were observed in all four altitudinal ranges. The findings also indicated that the higher the altitude is, the lower the Cr concentration, and vice versa. However, the Pb concentration was relatively high at high altitudes and relatively low at very low altitudes. In the case of Cd and As (purple and green bars, respectively), the concentration of these metals was greater at very low altitudes (Fig. 5).

Accumulation of heavy metals in soils associated with rice cultivation by agronomic management

The Pb concentration during agronomic management was highest at the phenological stage of filling (S3), reaching 12.5 mg kg^{-1} Pb (Fig. 6a), and the lowest concentration was obtained at the phenological stage of seedling (S1), at 9 mg kg^{-1}. The concentrations of As, Cd, and Cr were relatively high under the stubble burning cultivation technique, with values of 2, 2, and 16 mg kg^{-1}, respectively (Fig. 6b, 6c and 6 d, respectively). These values may be due to the presence of these metals in the leaf area of the plants, which can infiltrate into the soil when burned.

Distribution of heavy metals in soils related to rice cultivation via data interpolation

High As concentrations were observed in the northeastern part of the study area, with values ranging from 1.19 to 1.59 mg kg^{-1} (red area), whereas in the southwestern part of the study area, the concentrations were lower than 0.19 mg kg^{-1} (green area) (Fig. 7a). With respect to the distribution of Cd, this metal showed high concentrations in the northeastern part, with values between 1.4 and 1.7 mg kg^{-1} (red area), whereas in the southwestern part of the study area, values lower than 0.34 mg kg^{-1} were obtained (Fig. 7b).

The highest Cr concentration was observed in the northeastern part of the study area, and some areas in the western part exhibited high concentrations, with values ranging from 10.8 to 11.1 mg kg^{-1}. The lowest content of this metal was observed in the green area, with values between 10.2 and 10.6 mg kg^{-1} (Fig. 7c). The concentrations of Pb were higher in the central and western parts of the study area, with values ranging from 11.9 to 14.0 mg kg^{-1}. The northeastern part exhibited low Pb contents, with values ranging from 7 to 8.8 mg kg^{-1}, represented by the green area (Fig. 7d).

Analysis of the correlation coefficients between physicochemical parameters and heavy metals

A correlation coefficient diagram of the physicochemical variables and heavy metals revealed significant correlations ($p < 0.01$). The concentrations of OM and OC exhibited a perfect positive correlation (coefficient = 1); metals such as Cd and As also exhibited significant positive correlations (coefficient = 0.90), which suggests that with increasing Cd concentration, the As concentration also increased. In addition, the EC values of As and Cd were positively correlated, with correlation values of 0.36 and 0.42, respectively. However, the negative

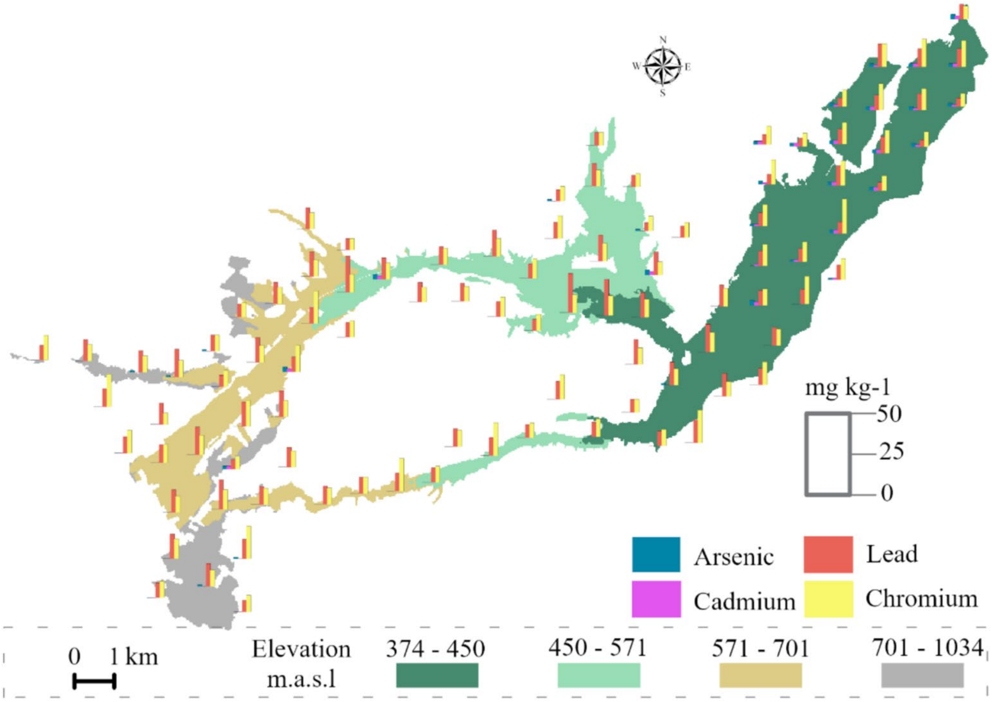


Fig. 5. Soil contaminant heavy metal distribution map (As: blue; Pb: red; Cd: violet; and Cr: yellow). The four elevation ranges are marked in different colors (dark green = very low; light green = low; mustard = medium; and gray = high).

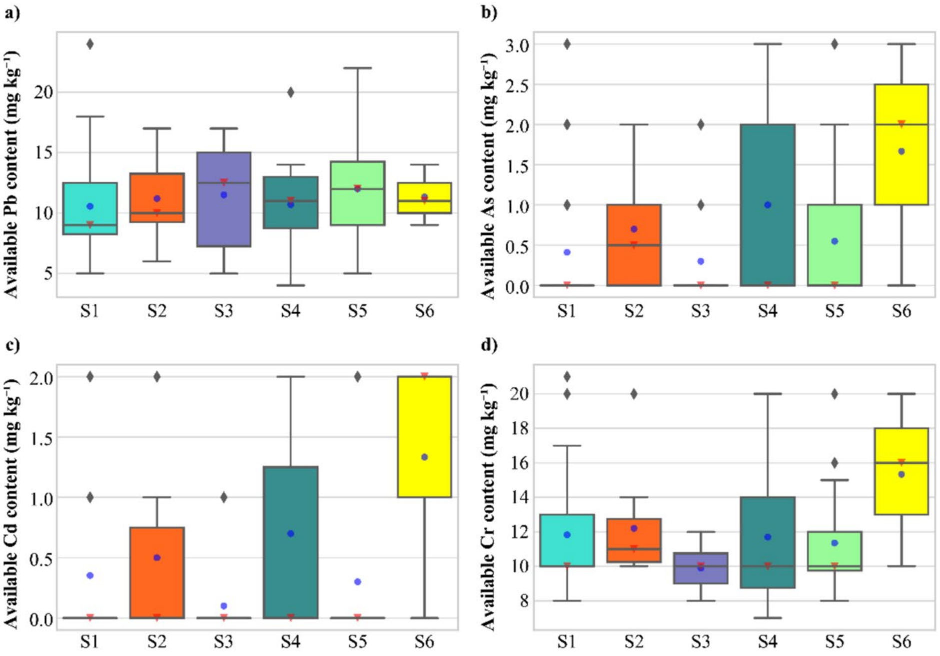


Fig. 6. Concentration of heavy metals at four phenological stages and under two rice cultivation techniques. Available contents of (**a**) Pb, (**b**) As, (**c**) Cd and (**d**) Cr. The four phenological stages of rice are S1 (seedling), S2 (tillering), S3 (filling), S4 (ripening), S5 (resting) and S6 (burning).

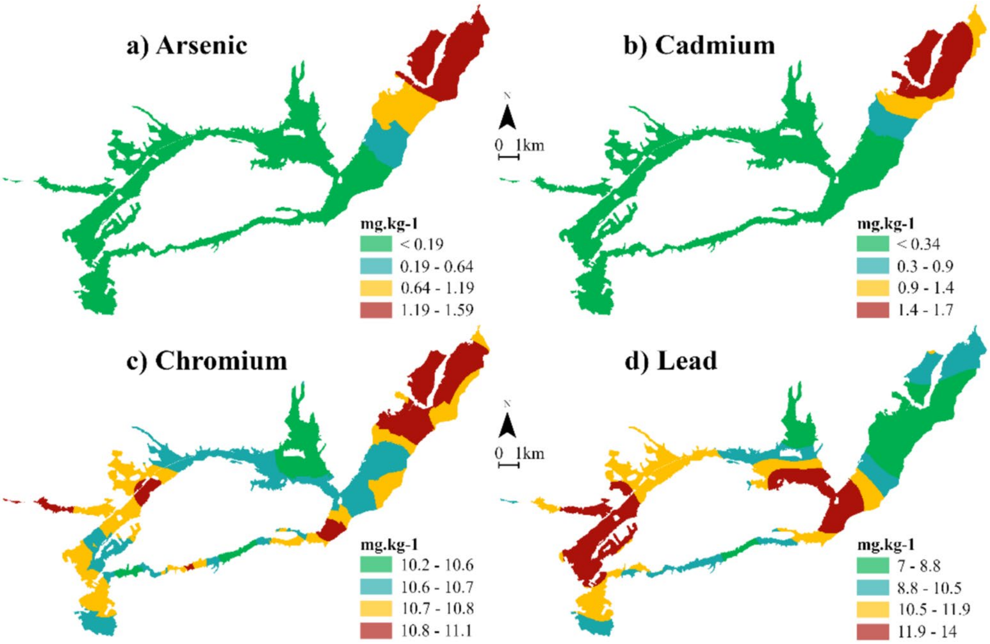
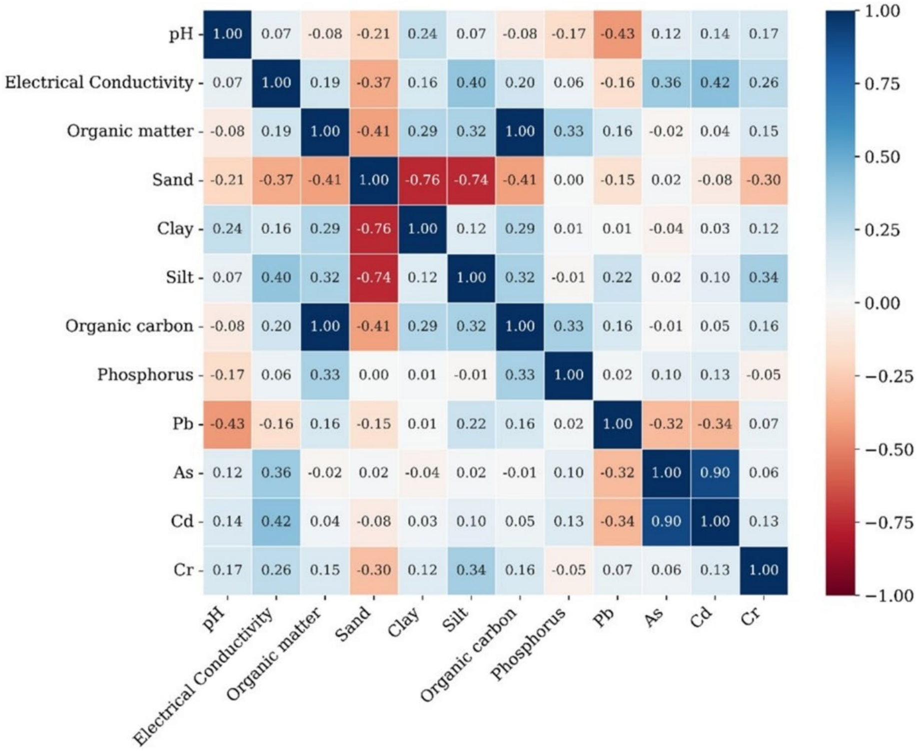


Fig. 7. Distribution of heavy metals via data interpolation. Four concentration intervals were employed (low concentrations: green; medium–medium high concentrations: light blue and yellow; high concentrations: red).

correlation between Pb and pH was highly significant, indicating that at lower pH values, the Pb concentration increased (Fig. 8).

Probability of heavy metal occurrence

The distribution probabilities indicated that the concentration of As was greater in the northeastern part of the study area, with values of up to 2 mg kg^{-1}. However, the concentration in the northern part decreased. Similarly,



	pH	Electrical Conductivity	Organic matter	Sand	Clay	Silt	Organic carbon	Phosphorus	Pb	As	Cd	Cr
pH	1.00	0.07	-0.08	-0.21	0.24	0.07	-0.08	-0.17	-0.43	0.12	0.14	0.17
Electrical Conductivity	0.07	1.00	0.19	-0.37	0.16	0.40	0.20	0.06	-0.16	0.36	0.42	0.26
Organic matter	-0.08	0.19	1.00	-0.41	0.29	0.32	1.00	0.33	0.16	-0.02	0.04	0.15
Sand	-0.21	-0.37	-0.41	1.00	-0.76	-0.74	-0.41	0.00	-0.15	0.02	-0.08	-0.30
Clay	0.24	0.16	0.29	-0.76	1.00	0.12	0.29	0.01	0.01	-0.04	0.03	0.12
Silt	0.07	0.40	0.32	-0.74	0.12	1.00	0.32	-0.01	0.22	0.02	0.10	0.34
Organic carbon	-0.08	0.20	1.00	-0.41	0.29	0.32	1.00	0.33	0.16	-0.01	0.05	0.16
Phosphorus	-0.17	0.06	0.33	0.00	0.01	-0.01	0.33	1.00	0.02	0.10	0.13	-0.05
Pb	-0.43	-0.16	0.16	-0.15	0.01	0.22	0.16	0.02	1.00	-0.32	-0.34	0.07
As	0.12	0.36	-0.02	0.02	-0.04	0.02	-0.01	0.10	-0.32	1.00	0.90	0.06
Cd	0.14	0.42	0.04	-0.08	0.03	0.10	0.05	0.13	-0.34	0.90	1.00	0.13
Cr	0.17	0.26	0.15	-0.30	0.12	0.34	0.16	-0.05	0.07	0.06	0.13	1.00

Fig. 8. Spearman's correlation coefficients of the soil physicochemical variables (pH, EC, OM, sand, clay, silt, OC and P) and heavy metals (Pb, As, Cd and Cr) with a categorized scale from 1–1 in red-blue, where red denotes the lowest correlation values from 0 to −1, and blue denotes the highest correlation values from 0 to 1.

the probability of Cd occurrence was higher in the northeastern part of the study area, with values of up to 2 mg kg^{-1}, whereas in the western part, the probability of the occurrence of this element was lower (Fig. 9).

The probabilities of Cr occurrence were greater in the northeastern part of the study area, whereas in the western part of the study area, the probability was lower. Moreover, a lower probability was obtained in the northern part of the study area. However, the probabilities of Pb occurrence were greater in the western part of the study area, whereas in the northeastern part, there was a lower probability of the occurrence of this metal.

Estimation of the standard error of the interpolation method

In the estimation of the standard error of the EBK method in the Amojú River basin (Fig. 10), the distributions of As (Fig. 10a) and Cd (Fig. 10b) revealed a pattern with higher standard errors in the northeastern part of the area, with standard error values ranging from 0.02 to 0.5, whereas the southwestern part exhibited lower standard errors, thus indicating high interpolation reliability. In contrast, Cr (Fig. 10c) and Pb (Fig. 10d) exhibited no distinct distribution patterns of the standard errors across the Amojú River basin, with values ranging from 0.7 to 1.1 for Cr and from 1.5 to 2.5 for Pb. These results indicated greater variability because the concentrations of these heavy metals were higher at select points in the Amojú River basin.

Calculation of the error metrics R^2, d, MAE, and RMSE of the EBK interpolation method

On the basis of the calculation of error metrics, the R^2 and d values were 0.7492, accounting for 74.9% of the explained variability in the data, indicating a favorable variability compared with that in other studies with a lower range of variability explained via the EBK method. Likewise, the analysis of the error metrics also revealed 25.1% of unexplained variability, which remains within a normal range of unexplained data. Moreover, the MAE value was 1.6615, and the RMSE value was 2.9075 due to the occurrence of moderate errors and extreme points that mostly affect the RMSE rather than the MAE value, as shown in Fig. 11.

The blue lines in Fig. 11a denote the observed values, and the red lines denote the predicted values. In Fig. 11b, the light blue dots denote the distributions of all the predicted and observed values in the study, and the green line denote the ideal range for the distribution of data via EBK interpolation.

Discussion

The soil pH is considered the main factor influencing the availability of metals in soil[34,35]. At high pH levels, metals commonly form barely soluble phosphates and carbonates. In contrast, at low pH levels, metals are likely to exist in more bioavailable free ionic forms[36]. In this study, the soils in the Amojú River basin exhibited an average pH of 7.8, indicating slightly alkaline conditions. Other studies of rice soils have revealed lower average

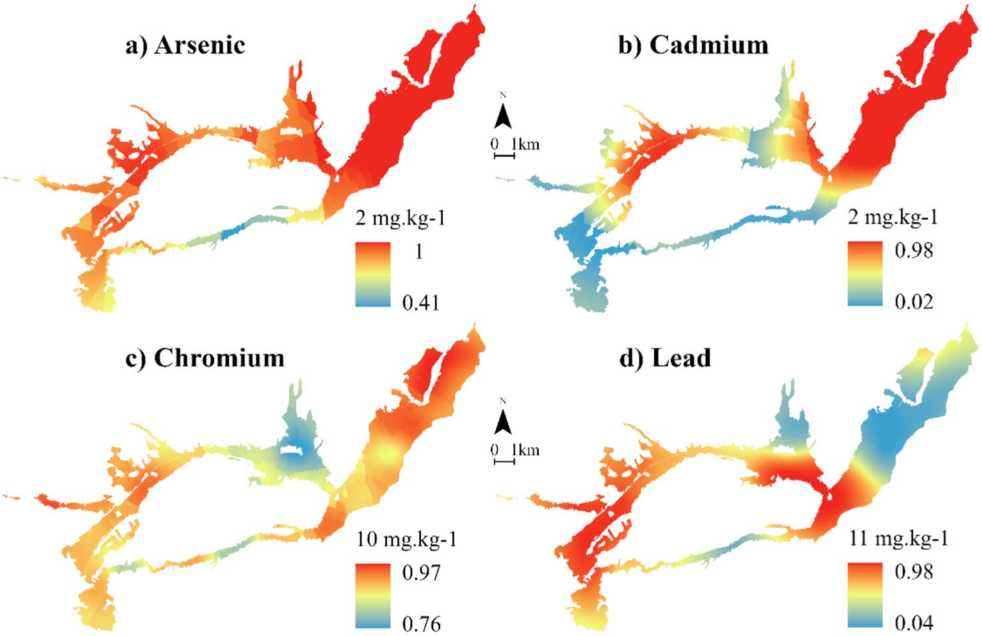


Fig. 9. Distribution probabilities of the occurrence of four heavy metals in the study area. The red areas indicate regions with a relatively high concentration of a given element, and the light blue areas indicate regions with a low concentration of the element.

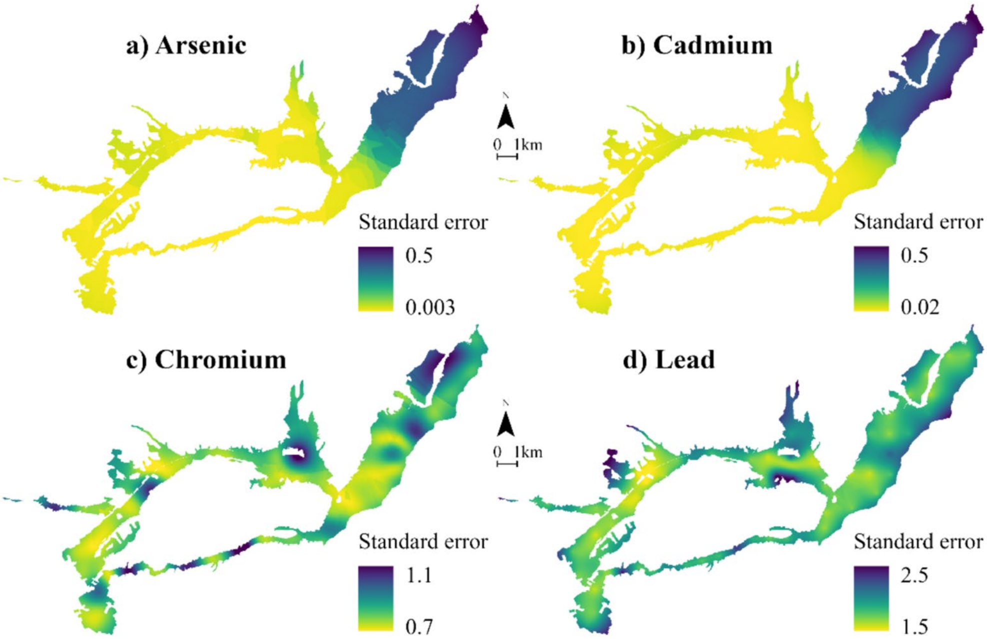
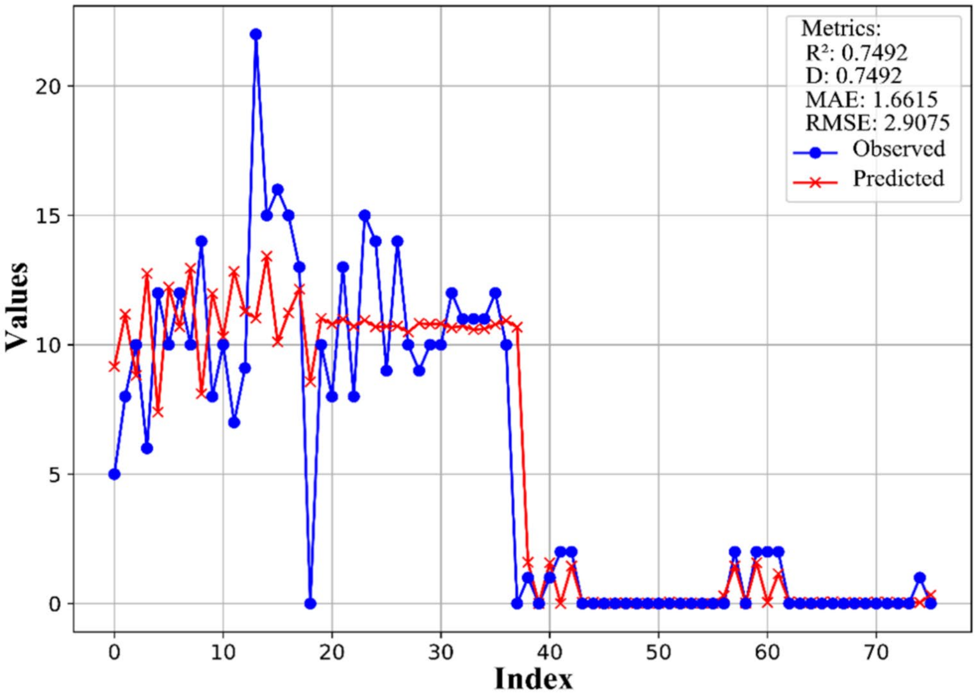


Fig. 10. Standard error estimation according to the empirical Bayesian kriging (EBK) method, in which the distributions of the heavy metals (**a**) AS, (**b**) Cd, (**c**) Cr and (**d**) Pb in the Amojú River basin are categorized by yellow–blue colors, where yellow denotes small standard errors, green denotes intermediate values, and blue denotes large standard errors according to the distribution of points in the Amojú River basin.

pH values (5.29)[37]. The behavior of heavy metals in soil can be predicted on the basis of their physicochemical properties[38]. Notably, the pH is a key factor because of its significant influence on biogeochemical processes[39].

Owing to its nature, OM is formed and distributed in different ways[40]; OM can function as a causative factor for the release of heavy metals but can also immobilize them[41]. The mean OM content in our study was 1.9%, indicating that its occurrence might be insufficient for heavy metal retention[42].

a) Metrics



b) Predicted vs Observed

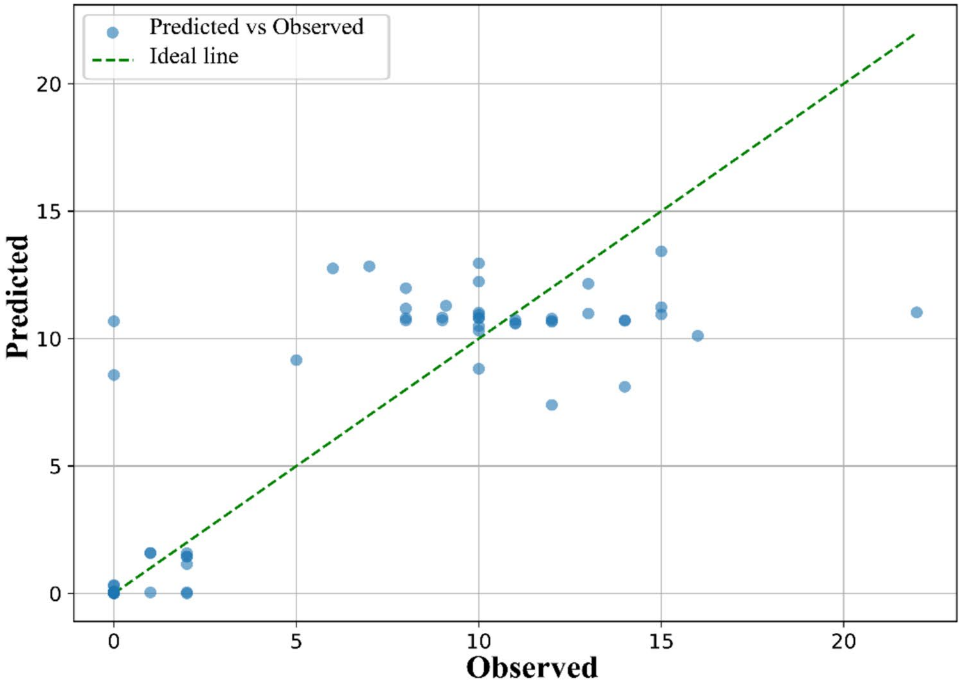


Fig. 11. Error metrics R^2, d, MAE, and RMSE, where subfigure (**a**) shows the metrics (blue = observed; red = predicted); and subfigure (**b**) shows the predicted vs. observed contents of heavy metals in the Amojú River basin (green diagonal line = ideal data distribution line; blue dots = predicted and observed data).

The EC is an indicator of soil salinity; in excess, it is detrimental to plant development, affecting growth and physiological parameters such as hormone production[43]. The EC of the rice-growing soils in the Amojú River basin averaged 21.8 mS m^{-1}, which is an indicator of the presence of saline soils in the study area. Soils with EC values higher than 0.8 mS m^{-1} are considered severely saline[34].

P is an essential macronutrient for plants, but it is also the most limiting factor for plant production in areas with tropical climates[44]. For this reason, available P concentrations between 4.4 and 65.5 mg kg^{-1} in soils associated with rice cultivation were obtained in this study. With respect to soil-pollutant interactions, clay soils have been the most extensively studied[34]. P availability for plants is related to the soil pH, considering that in very acidic soils, P is bound to Fe and Al oxides, thus reducing their availability to plants[45]. Our study revealed differences between soil textures in terms of the sand, clay and silt contents, indicating the presence of clay loam soils (Table 4).

Floodplain areas generally exhibit a homogeneous distribution of heavy metal contamination[46]. In our study area, which corresponds to the very low altitudinal range (374–450 m a.s.l.), a higher Cr concentration was obtained, with a median of 12 mg kg^{-1}. Lower concentrations of Cd, Pb, and As were observed, with median values of 1, 9, and 1.5 mg kg^{-1}, respectively. Similar results were obtained in a study conducted in soils with rice crops, where high concentrations of Cd (> 0.47 mg kg^{-1}), Pb (> 49.9 mg kg^{-1}) and Cr (93.7 mg kg^{-1}) were observed at altitudes between 30 and 60 m a.s.l[12].. Similarly, in several studies, the presence of heavy metals such as As, Pb, Cr and Cd in agricultural soils has been attributed to the intensive use of phosphorous fertilizers[14,27].

The presence of heavy metals poses a risk to both the environment and human health[47,48]. The World Health Organization (WHO) has identified Cd as a toxic heavy metal that poses a significant risk to human health, and its accumulation in rice agricultural soils may represent toxicity[49]. According to the Environmental Quality Standards (EQSs), the allowable limit for the Cd concentration in agricultural soils is 1.4 mg kg^{-1}. However, in this study, up to 2 mg kg^{-1} of Cd was obtained (17% of the sampling points), a value that exceeds the EPA limits. Similar results were reported for soils associated with rice cultivation in Venezuela, with Cd concentrations higher than 2.26 mg kg^{-1}[50].

Elevation and slope could be among the main factors influencing heavy metal enrichment[51,52]. In our study, the Cd concentrations were relatively high within the very low altitudinal range (374 to 450 m a.s.l.), possibly because of crop irrigation wastewater. Similar results were obtained in a study conducted in agricultural soils, which revealed that the Cd concentration increased by 52% as the elevation decreased. This behavior was compared between areas below 2,100 m and areas above 2,250 m. In addition, the As and Pb contents in upland areas were significantly higher than those at lower elevation sites[51].

Notably, high contents of As, Cd and Cr have been obtained when the soil has undergone a recent process of vegetation burning, possibly because, during the burning of vegetation and wood, some elements are not volatilized but are contained in the ashes, which can leach into the soil profile and accumulate[53]. This accumulation of heavy metals in agricultural land can directly affect the growth and productivity of plants[37].

The correlation between the distributions of Cd and As, with similar concentrations and distributions throughout the study area, revealed that the northeastern part exhibits the highest concentrations, with values of > 1.4 mg kg^{-1} for Cd and > 1.19 mg kg^{-1} for As, which could be due mainly to the accumulation of waste substances in irrigation and the abuse of phosphorus fertilizers in rice cultivation[14]. Similar to studies conducted on the old continent, the distributions of both metals reveal similar values, with values of > 0.56 mg kg^{-1} for Cd and > 40.00 mg kg^{-1} for As[1].

Future research should not only focus on soil analysis but also focus on root, leaf and grain analyses of rice and other transient, permanent and rotational crops to develop soil remediation strategies, such as the use of biochar or phytoremediation measures, microorganisms and phytoremediation plants, to strengthen environmental policies and sustainable agricultural practices.

Conclusions

Heavy metal contamination of soils associated with rice cultivation in the Amojú River basin is a challenge that must be addressed urgently. This study revealed that Cd concentrations exceeded EQSs (> 1.4 mg kg^{-1}) at 17% of the sampling points. Similarly, the highest concentrations of Cr and Cd were obtained in the lower-altitude areas (374–450 m a.s.l.) This could be related to the intensive excessive use of pesticides and/or phosphorus fertilizers and/or pesticides for crop development.

Notably, Cd, As, and Cr, with concentrations of 2, 2, and 16 mg kg^{-1}, respectively, were detected in areas where stubble burning is practiced, which suggests that this common traditional agricultural activity could contribute significantly to the accumulation of these metals in the soil within this area. These findings also suggest the possible mobilization of heavy metals into the leaf area of the foliar tissues of cultivated plants.

The results of this study provide an initial overview of heavy metal contamination in agricultural soils, which can help local, regional, and national authorities implement management and monitoring strategies for As, Cd, and Pb, as they may pose a risk to human health.

One of the limitations of this study was the restricted access to certain rice plots because some farmers did not authorize soil sampling. This may have reduced the spatial representativeness of sampling and limited the coverage in specific productive areas. Therefore, the integration of entities for interinstitutional cooperation between farmers and institutions is recommended to bridge the gap when research involving sampling in private agricultural areas is needed. In addition, proponse complementary research on contamination sources, the traceability of chemicals and the risks to human health should be assessed to better understand the mechanisms of contamination and design effective mitigation measures.

It is recommended that complementary research be conducted that includes the direct analysis of agricultural inputs, monitoring of contaminants in plants and water sources, and human health risk assessment to better understand the mechanisms of contamination and design effective mitigation actions.

Data availability

The datasets generated and/or analyzed in this study are provided in the Supplementary Information files.

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Author contributions

E.T: Research, Methodology, Writing—original draft,; M.A.I: Conceptualization, Formal Analysis, Visualization; P.T.H: Data Curation, Methodology; D.T: Formal Analysis, Validation; V.T.M: Financial Procurement, Project Management; J.C.L: Financial Acquisition, Resources; N.R.B: Data Curation, Writing; N.A.M: Data Curation, Validation; D.G.F: Data Curation, Methodology, Validation; M.G: Conceptualization, Formal Analysis, Validation.

Declarations

Competing interests

The authors declare no competing interests.

Additional information

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